



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of Tatsuya Yoshikawa

Serial No.: 09/888,009

5 Filed: June 22, 2001

Title: EXHAUST GAS PURIFYING CATALYST

Art Unit: 1754

Examiner: JONASAN STRICKLAND

10 COMMISSIONER FOR PATENTS

WASHINGTON, D. C. 20231

DECLARATION UNDER 37 C. F. R. 1.132

Sir:

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I. I, Tatsuya YOSHIKAWA, of B-612, 1736-3, Imajuku, Himeji, Hyogo 670-0052, Japan, hereby declare that I am an inventor named in the above-mentioned application, that I graduated from Kyoto University, Department of Engineering, Petroleum Chemistry, in March 1988 with a B.S. degree. I received my master's degree, molecular engineering, from the same University in March 1990, that, since April 1990, I have been employed by Nippon Shokubai Co., Ltd., and engaged in exhaust gas purifying catalyst research for last 7 years. In the
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25 period, I have been to Reading University, UK, for 2 years as guest researcher to study surface science from 1994 to 1996. Additionally, in accordance with our section consolidation to ICT Co., Ltd, I have been in temporal transfer to ICT Co., since 1998.

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II. I understand that claims 13-15 of the above application has been rejected under 35 U. S. C. 103 (a) as being unpatentable

over Kato et al. (EP 0365308A2) in view of Montreuil et al. (US Patent 5,328,672) and I have reviewed these references. It is of my opinion that the claimed catalysts in the present application are nonobvious over the cited references because
5 the claimed catalyst achieved superior performance as shown by the following experimental results.

10 EXPERIMENT 1 (corresponding to Example 1 of the present specification)

One thousand (1000) g of zirconium oxide was impregnated with an aqueous solution containing a 100 g equivalent of manganese nitrate as manganese oxide (MnO_2), dried overnight at 120°C, and calcined at 550°C for one hour. The resultant
15 manganese-deposited zirconium oxide and H-ZSM5 (Si/Al = 27, available from Zeolyst International) were separately ball milled in an aqueous solution, and mixed at a weight ratio (solid ratio) of 2: 8. The produced mixture was applied to a monolith made of cordierite (400 cells, 0.043 liter,
20 available from NGK Insulators, LTD in Japan) so as to account for an amount of 180 g/L. The coated monolith thus obtained was dried at 120°C and then calcined at 500°C for one hour to obtain a catalyst. The catalyst performance was measured under the conditions in Table1.

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EXPERIMENT 2 (corresponding to Example 2 of the present specification)

One thousand (1000) g of zirconium oxide was impregnated with an aqueous solution containing a 100 g equivalent of
30 manganese nitrate as manganese oxide (MnO_2) and a 40 g equivalent of cerium nitrate as cerium oxide (CeO_2), dried overnight at 120°C, and calcined at 550°C for one hour. The

resultant manganese-deposited zirconium oxide and HZSM5 (Si/Al = 27, available from Zeolyst International) were separately ball milled in an aqueous solution, and mixed at a weight ratio (solid ratio) of 2: 8. The produced mixture was applied to a monolith made of cordierite (400 cells, 0.043 liter, available from NGK Insulators, LTD in Japan) so as to account for an amount of 180 g/L. The coated monolith thus obtained was dried at 120°C and then calcined at 500°C for one hour to obtain a catalyst. The catalyst performance was measured under the conditions in Table1.

In the reaction gas shown in Table 1, the produced catalyst was heated from 100°C to 550°C at an increasing rate of 20°C/min. and cooled from 550°C to 150°C at a decreasing rate of 20°C/min. to determine conversion efficiencies of NO_x.

TABLE 1. CONDITIONS OF REACTION GAS

| Conditions for rating model gas | |
|---|--|
| NO: 400 ppm, | |
| Propylene: 1800 ppm C ₁ (normalized to carbon concentration), CO: 400 ppm, O ₂ : 8.5 vol %, Water: 10 vol % | |
| Total flow volume: 28 L/min., SV: About 40,000 h ⁻¹ | |

NO was analyzed with a chemical emission spectral analyzer (CLA), and the hydrocarbon with a hydrogen flame ionizing analyzer (FIA), respectively.

The results obtained are shown in Table A below.

COMPARATIVE EXPERIMENT 1

A catalyst was prepared according to the procedure of

Experiment 1 while using Cu-Mordenite (Example 1 in Kato et al.) instead of H-ZSM5. The catalyst performance was measured under the conditions in Table 1 and the result obtained is shown in Table A below.

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COMPARATIVE EXPERIMENT 2

A catalyst was prepared according to the procedure of Experiment 1 while using Cu-ZSM5 (Example 1 in Montreuil et al.) instead of H-ZSM5 and using CuOx/ZrO₂ (Example 1 in Montreuil et al. and also Kato et al.) instead of MnOx/ZrO₂. Cu content in the CuOx/ZrO₂ powder was 10wt% as CuO. The catalyst performance was measured under the conditions in Table 1 and the result obtained is shown in Table A below.

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COMPARATIVE EXPERIMENT 3

A catalyst was prepared according to the procedure of Experiment 2 while using Cu-ZSM5 instead of H-ZSM5. The catalyst performance was measured under the conditions in Table 1 and the result obtained is shown in Table A below.

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TABLE A

| | Catalyst | | NOx conv. (%) |
|--------------------------|-------------------------|---------------|---------------|
| | 1st Component | 2nd Component | HC/NOx=4.5 |
| Experiment 1 | MnOx/ZrO ₂ | H-ZSM5 | 55 |
| Experiment 2 | MnCeOx/ZrO ₂ | H-ZSM5 | 75 |
| Comparative Experiment 1 | MnOx/ZrO ₂ | Cu-Mordenite | 28 |
| Comparative Experiment 2 | CuOx/ZrO ₂ | Cu-ZSM5 | 36 |
| Comparative Experiment 3 | MnCeOx/ZrO ₂ | Cu-ZSM5 | 32 |

Note: Maximum NOx conv. was determined in the temp. descending test.

CONCLUSION

5 It is clear from the above Table A that the catalyst of the present invention provided superior NOx conversion to the catalysts of the Comparative Experiments 1 and 2, which are derived from Kato et al. and Montreuil et al and also of the Comparative Experiment 3.

10 III. The undersigned declares that all statements made herein are true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fines or imprisonment, or both under
15 Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

20 July 16th, 2003
Date

Tatsuya Yoshikawa
Tatsuya YOSHIKAWA